New mechanism for exciton-biexciton changes in the spectra of a semiconductor in a polariton pump wave

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(Submitted 25 November 1988)
Pis'ma Zh. Eksp. Teor. Fiz. **49**, No. 1, 34–36 (10 January 1989)

A new mechanism for the exciton-biexciton changes in the spectra of a semiconductor is proposed. This mechanism operates because of a direct virtual binding of two excitons to form a biexciton by virtue of their Coulomb interaction. It is shown that this mechanism determines the structural changes in spectra which have previously been linked with exclusively a "giant" oscillator strength of the exciton-biexciton transition. In particular, this new mechanism leads to an effective long-wavelength shift of exciton and biexciton levels.

Exciton-biexciton changes in the spectra of a semiconductor in a polariton pump wave were first studied in Ref. 1–3. This effect was originally linked with a giant oscillator strength of an exciton-biexciton transition.[4,5] Formally, that explanation implied incorporating terms of the type $(1/\sqrt{V})M_1(\mathbf{p},\mathbf{q})A_{\mathbf{p}}^{+}B_{\mathbf{q}}\alpha_{\mathbf{p}-\mathbf{q}}$, in the phenomenological Hamiltonian under consideration, where $A_{\mathbf{p}}$, $B_{\mathbf{q}}$, and $\alpha_{\mathbf{p}-\mathbf{q}}$ are operators which annihilate a biexciton, an exciton, and a photon, respectively, and are introduced in an independent way. In other words, only the process in which a biexciton $\mathbf{p}$ is created through the absorption by exciton $\mathbf{q}$ of a photon $\mathbf{p}-\mathbf{q}$ is taken into account. However, another process, which is determined by the term $(1/\sqrt{V})M_2(\mathbf{p},\mathbf{q})A_{\mathbf{p}}^{+}B_{\mathbf{q}}B_{\mathbf{p}-\mathbf{q}}$, correspondingly, is physical. It describes a direct binding of two excitons, $\mathbf{p}-\mathbf{q}$ and $\mathbf{q}$, into an exciton $\mathbf{p}$ by virtue of their Coulomb attraction. It is understood implicitly that there are third quasiparticles—photons—which satisfy the energy conservation law in processes of this sort. Roughly speaking, it is the polariton effect which makes possible a direct binding of two excitons to form a biexciton. Furthermore, we will show that this mechanism is in fact responsible for the changes in the spectra and that it leads to several qualitatively new results.

Our model Hamiltonian for the photon-exciton-biexciton system is constructed

 0021-3640/89/010039-04$01.00

on the basis of the approximation by the operator for $H = H(A, B, \alpha)$ of the original elementary Hamiltonian, which describes electrons and holes that are interacting with each other in accordance with Coulomb's law. These particles are also in an electromagnetic field, which causes band-band electron transitions. An important point is that a biexciton is implicitly assumed to consist of two excitons in the quasiparticle Hamiltonian found; i.e., we have

$$A_{\mathbf{p}} = \frac{1}{\sqrt{2V}} \sum_{\mathbf{l}} \psi(\mathbf{l}) B_{-\mathbf{l}+\frac{\mathbf{p}}{2}} B_{\mathbf{l}+\frac{\mathbf{p}}{2}}, \tag{1}$$

where $\psi(\mathbf{l})$ is the wave function of the relative motion of the excitons in the biexciton, and V is the volume of the crystal. This approximation corresponds to the case in which the exciton binding energy is substantially greater than the binding energy of the corresponding biexciton.

The changes in the spectra of elementary excitations in a semiconductor in an intense, coherent, polariton wave $\mathbf{k}$ of frequency $\omega_{\mathbf{k}}$, which lies in the transparency region near the exciton absorption line, is described by replacing the exciton and photon operators of the selected mode by C-numbers: $B_{\mathbf{k}} \to \sqrt{V} P \exp(-i\omega_{\mathbf{k}} t)$, $\alpha_{\mathbf{k}} \to \sqrt{V}\mathcal{E} \exp(-i\omega_{\mathbf{k}} t)$, which are actually exciton and photon components of the pump wave. In this case the quasiparticle Hamiltonian in which we are interested takes the following form after a simple canonical transformation which eliminates the explicit time dependence:

$$H = \sum_{\mathbf{p}\neq\mathbf{k}} \{(\omega_{\mathbf{p}}^{ph} - \omega_{\mathbf{k}})\alpha_{\mathbf{p}}^{+}\alpha_{\mathbf{p}} + (\omega_{\mathbf{p}}^{ex} + 2\epsilon_{\mathbf{p}+\mathbf{k}} | \psi(\frac{\mathbf{p}-\mathbf{k}}{2})|^2 P^* P - \omega_{\mathbf{k}}) B_{\mathbf{p}}^{+} B_{\mathbf{p}}$$
$$+ \epsilon_{\mathbf{p}+\mathbf{k}} A_{\mathbf{p}+\mathbf{k}}^{+} A_{\mathbf{p}+\mathbf{k}}\} + \sum_{\mathbf{p}\neq\mathbf{k}} \{\sqrt{2}\epsilon_{\mathbf{p}+\mathbf{k}} \psi(\frac{\mathbf{p}-\mathbf{k}}{2}) P A_{\mathbf{p}+\mathbf{k}}^{+} B_{\mathbf{p}} - i\frac{\Omega_c}{2} B_{\mathbf{p}}^{+}\alpha_{\mathbf{p}} + h.c.\}, \tag{2}$$

where $\omega_{\mathbf{p}}^{ex}$ ($\omega_{\mathbf{p}}^{ph}$) is the unperturbed dispersion of the excitons (or photons), the parameter Ω_c is a measure of the strength of the exciton-photon interaction (i.e., the ordinary polariton effect), and the quantity $\epsilon_{\mathbf{p}}$ is defined by

$$\epsilon_{\mathbf{p}} = \Omega_{\mathbf{p}}^{biex} - \frac{1}{V} \sum_{\mathbf{q}} [\omega_{-\mathbf{q}+\frac{\mathbf{p}}{2}}^{ex} + \omega_{\mathbf{q}+\frac{\mathbf{p}}{2}}^{ex}] | \psi(\mathbf{q}) |^2. \tag{3}$$

The systematic use of the microscopic approach in the construction of the model Hamiltonian leads to several substantial distinctions from the conventional theories.[1-3] In the first place, the exciton and biexciton excitations can no longer be treated as independent, as follows from (1):

$$[A_{\mathbf{p}}, B_{\mathbf{q}}^{+}] = \sqrt{\frac{2}{V}} \psi(\mathbf{q} - \frac{\mathbf{p}}{2}) B_{\mathbf{p}-\mathbf{q}} \tag{4}$$

Second, the quantity $\epsilon_{\mathbf{p},\mathbf{k}}$ in Hamiltonian (2) is not equal to the unperturbed biexciton energy $\Omega_{\mathbf{p},\mathbf{k}}^{biex}$; it is determined by the average potential energy of the interaction of the electrons in the biexciton, (3). Furthermore, matrix element $M_1(\mathbf{p}, \mathbf{q})$ and the

terms associated with it are completely missing from (2). Hamiltonian (2) is, as before, a measure of the giant oscillator strength of the exciton-biexciton transition, which is now determined in a natural way by the form of biexciton operator (1) and by the polariton parameter Ω_c (in particular, the matrix element of the two-photon transition to the biexciton state is the same as that calculated in Ref. 9). We should point out here that the error in the approximation of the original elementary Hamiltonian and, in particular, the relative error in the determination of the parameter ϵ_p are on the order of $(a_{ex}/a_{biex})^3 \ll 1$, where a_{ex} and a_{biex} are the first Bohr radii of the exciton and the biexciton, respectively. Furthermore, in determining the parameters of Hamiltonian (2) we did not use corrections on the order of $N_{ex}a^3_{biex}$, so the results derived here can be applied only under the condition $Ia^3_{biex}/\hbar\omega_k v_k \ll 1$, where I is the pump intensity, and v_k is the corresponding polariton group velocity.

By putting quadratic Hamiltonian (2) in diagonal form through the introduction of some new elementary excitations $\tilde{A}_{p+k}$, $\tilde{B}_p$, and $\tilde{\alpha}_p$, which are expressed in a linear fashion in terms of the quasiparticle operators A_{p+k}, B_p, and α_p, we can determine the reconstructed spectrum of the semiconductor in the presence of the pump wave. This procedure is determined in an unambiguous way by the requirement that the new excitations be of a boson nature and by the condition that they be independent, which means, in particular, $[\tilde{A}_{p+k}, \tilde{B}^+_p] = 0$. The most interesting results, in our opinion, are the shift of the exiciton and biexciton levels in the long-wave direction, which depend on the pump intensity. The effect is determined by the direct exciton-exciton interaction, which gives rise to the formation of biexcitons. Specifically, the shift $\Delta_{ex} = 2\epsilon_{p+k}|\psi[(\mathbf{p}-\mathbf{k})/2]|^2 P^* P$ of the exciton level in the red direction is of the

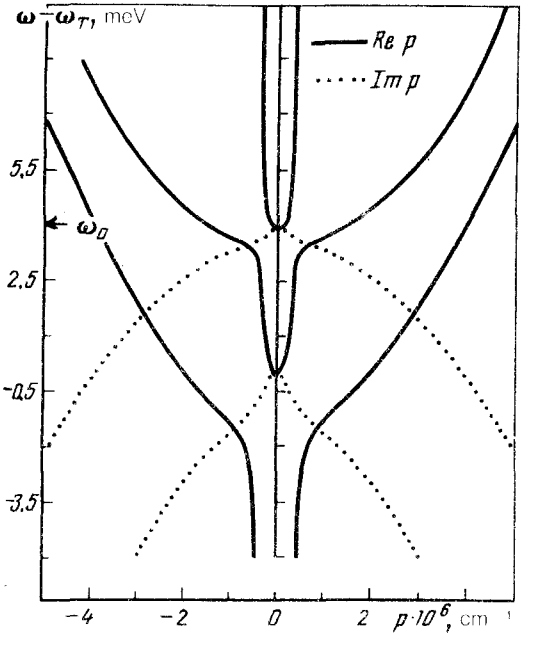

FIG. 1.

following nature. An exciton $\mathbf{p}$ experiences an attraction from the excitons $\mathbf{k}$ of the pump wave. This attraction causes a virtual formation of biexcitons. The effectiveness of this interaction is determined by the corresponding biexciton potential $\epsilon_{\mathbf{p}+\mathbf{k}} < 0$, while the quantity $|\psi[(\mathbf{p}-\mathbf{k})/2]|^2$ determines the probability for the virtual formation of a biexciton from excitons $\mathbf{p}$ and $\mathbf{k}$. An important point is that the nature of the interaction of the excitons usually allows us to assume $\psi[(\mathbf{p}-\mathbf{k})/2] \approx \psi(0)$, i.e., to treat the shift of the exciton level as a whole in the optical region, $p \lesssim 10^6$ cm^{-1}, in which we are interested. The nature of the biexciton shift $\Delta_{\text{biex}} \approx 2\Delta_{\text{ex}}$, which arises is similar; the existence of such a shift has been indicated by experiments.[6] This level-shift effect is of a dynamic nature and in this sense completely analogous to the optical Stark effect,[7] but—an important point—the thresholds for the observation of this effect in terms of the pump intensity are several orders of magnitude lower. With regard to the exciton-biexciton splitting at the frequency $\omega_0 = \Omega_0^{biex} - \omega_{\mathbf{k}}$, we note that it turns out to be substantially smaller than was predicted in Refs. 1–3, since it is determined not by the giant oscillator strength but by the matrix element $\tilde{M}_2 = \sqrt{2}\epsilon_{\mathbf{p}+\mathbf{k}}\psi[(\mathbf{p}-\mathbf{k})/2]P$. This situation appears to be closer to the situation observed experimentally.[8]

Figure 1 shows dispersion curves of the reconstructed spectrum of the semiconductor CdS for the geometry $\mathbf{p}\perp\mathbf{k}$ which arise by virtue of this new interaction mechanism. For a numerical solution of the dispersion relation we used the following parameter values $\omega_T = 2552$ meV, $\Omega_0^{biex} = 5100$ meV, $\omega_{\mathbf{k}} = 2544$ meV, $I = 10$ MW/cm^2, $\epsilon = -5$ meV, $M_{\text{ex}} = 0.9m_0$, $M_{\text{biex}} = 2M_{\text{ex}}$, $\epsilon_g = 8.87$, and $|\psi(0)|^2 = 10^{-18}$ cm^3.

We wish to express our sincere gratitude to L. V. Keldysh for numerous discussions of these results.

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Translated by Dave Parsons